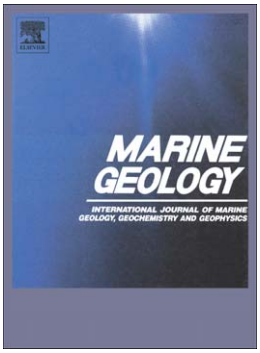

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Geochemical and Sr-Nd isotopic variations in a deep-sea sediment core from eastern Indian Ocean: Constraints on dust provenances, palaeoclimate and volcanic eruption history in the last 300,000 years

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Abstract: This study reported geochemical and Sr-Nd isotopic variation of fine-grained mineral components in a deep-sea piston core G6-4, south of the Sumba Island, Eastern Indian Ocean. The new data enables us to characterise dust contributions from Australian continent and volcanic eruption history in the adjacent volcanic arc over approximately the last 300,000 years. The geochemical and isotopic features of the fine-grained components (<20 μm) in Core G6-4 are broadly consistent with mixing between aeolian dusts from the Australian continent and volcanic ashes of the Sunda arcs. Simple two-component modeling shows that the majority of the core samples probably are composed of 50-80% aeolian dusts derived from the Australian continent. Sr-Nd isotopic ratios are fairly uniform for samples older than 180 ka, in contrast with those of the samples younger than 180 ka, which are much more variable with three anomalies characterized by higher $^{143}Nd/^{144}Nd$ and lower $^{87}Sr/^{86}Sr$ values recognized. We interpret the three anomalies in the last 80 ka as recording episodes of eruptions from nearby volcanoes. Thus, it is likely that volcanism become more active in the last 80 ka. Excluding these anomalies, the average $^{143}Nd/^{144}Nd$ was generally lower, and average $^{87}Sr/^{86}Sr$ higher in the post-180 ka section than in the older section, which might be associated with increased Australian continental aridity towards more recent time.

Keywords: Dust provenance; Palaeoclimate; Volcanic eruption; geochemistry; Sr-Nd isotopes; Eastern Indian Ocean; Late Quaternary

1. Introduction

Marine sediments, especially aeolian dusts, preserved in deep sea sediment cores provide valuable information on tracing dust provenance and continental climatic change (e.g. Rea and Janecek, 1981; Staudigel et al., 1995; Holm, 2002; Wei et al., 2004; Grousset and Biscaye, 2005; Bayon et al., 2009; De Deckker et al., 2010; Le Houedec et al., 2012).

As Australia is the driest inhabited continent on the planet, there has always been a lot of interest in its climate history. Previous studies (e.g. Bowler, 1976; Hesse and McTainsh, 2003; De Deckker et al., 2010) concluded that there are two general dust paths in the Australian region, namely towards the southeast and the northwest. The southeast dust path has received by far the most research attention (e.g. McTainsh, 1989; Hesse and McTainsh, 2003; Revel-Rolland et al., 2006; Marx et al., 2009; Petherick et al., 2009; De Deckker et al., 2010). However, with few exceptions (e.g. Stuut et al., 2014), the contribution of the aeolian dusts by the northwest dust path to the Indian Ocean was not investigated until comparatively recently, particularly during the late Quaternary period (e.g. Kohfeld and Harrison, 2001; Ehlert et al., 2011; McGowan and Soderholm, 2012), in part due to the paucity of long time-scale archives. Another non-negligible difficulty is associated with recognizing Australian dusts within sedimentary deposits of mixed origin (Gasparon and Varne, 1998) as aeolian dusts deposited in the Indian Ocean are generally well mixed with volcanic ashes (Gasparon and Varne, 1998; Gingele et al., 2001) and thus represent the average source signal. Difficulties are often met to estimate the respective contribution of

these two source components.

In this study, 25 fine-grained samples were selected from the sediments collected from a piston core G6-4 at a water depth of 3510m, which is situated in the monsoonal belt, south of the Sumba Island in the Indian Ocean, on the down-wind dust path from the Australian continent. Following previous investigations on fire history of the Australia continent and adjacent Islands over the past 300 ka (Wang et al., 1999), the present study is intended to use Sr-Nd isotopic signatures to characterise the provenance of the mineral components in the core. Since aeolian dusts from the Australia continent have isotopic compositions distinctive from those of volcanic ashes from the Sunda island arcs, the proportion of contributions from the Australia continent as comparing with volcanic ashes from the Sunda island arcs in the vicinity of the core can be readily estimated. The result will provide further constraints on monsoonal activity and climatic change over the past 300 ka.

The additional purpose of this study is to use Sr-Nd isotopic data derived from the core section to characterise specific volcanic eruption events in the adjacent volcanoes. Volcanic ashes from volcanoes nearby are expected to be preferentially indicated in the core. Since this core has a well-established marine oxygen isotope chronology (Wang et al., 1999), the timing of each volcanic eruption can also be estimated. The characterization of volcanic eruption events in the adjacent volcanic arcs has important implications for long-term volcanic hazard assessment. The study method has general application to dating eruption recurrence events elsewhere.

2. Core G6-4 and its regional setting

From June 1984 to July 1985, a joint Indonesian-Dutch marine scientific expedition, the Indonesian-Dutch Snelliusll expedition, was carried out in the eastern Indonesian water to reconstruct the vegetational and climatic history of the east Indonesian-north Australian area. Detailed descriptions of the Indonesian-Dutch SnelliusⅡ expedition and the techniques used for sampling are given by Van Hinte (1989) and Van der Kaars (1991). A deep-sea piston core (G6-4) in this study was obtained during the execution of the 1984-1985 Indonesian-Dutch SnelliusⅡ expedition. This core was taken at 10º47´S, 118º04´E, from the Lombok Ridge, approximately 170 km SW off Sumba and 850 km NW of northern Australia, at a water depth of 3510m (Fig.1).

The Lombok Ridge is part of the Sunda-Banda arc system which is built on crust whose thickness is transitional between continental and oceanic domains. The Sunda-Banda arc system forms the southern and southeastern margin of the general Indonesian region, one of the most tectonically active areas of the present Earth's surface (Foden and Varne, 1980). Volcanic activity in the arc is a response to northward dipping subduction of the Australian plate (Elburg et al., 2007). The active volcanic arc is marked by a line of active volcanoes running almost continuously from Sumatra to the eastern Banda Sea (Foden and Varne, 1980;Lytwyn et al., 2001)(Fig.1). Igneous rocks display a wide geochemical range from basalts and gabbros to andesites and diorites (Lytwyn et al., 2001). Previous geochemical studies of neighboring volcanic rocks along the eastern Sunda and Banda Arc systems are largely restricted

to Quaternary eruptions (Vroon et al., 1995). The active volcano on the Lombok Island is Rinjani, which is located approximately 165 km above the Benioff zone.

Approximately 900cm of sediment cores was recovered, mostly calcareous clays. There is clear evidence to demonstrate that the aeolian dusts emanating from Australia during Quaternary have produced significant deposits in the Indian Oceans (McTainsh, 1989; Hesse and McTainsh, 2003). Sediments have been supplied to Eastern Indian Ocean through northwest dust path-way (McTainsh, 1989). Pollen record from Core G6-4 indicated that approximately 80% of the pollen was derived from Australia (van der Kaars, 1991). Based on 90 oxygen dates of *Globigerinoides ruber* and 4 radiocarbon dates of foraminifera presented in Wang et al. (1999), the refinement of the oxygen isotope record has provided a firmer chronology for the palaeoenvironmental record (Wang et al., 1999). The age model in Wang et al. (1999) was determined by aligning characteristic features from the benthic $\delta^{18}O$ record with equivalent features in the stacked isotopic record of Martinson et al. (1987) using the software 'Analyseries' developed at the Centre des Faibles Radioactivitès (Paillard et al., 1996). The timescale was determined by correlation with the SPECMAP record (Martinson et al., 1987), and the ages of 25 samples in this study were linearly interpolated between control-points.

3. Analytical Methods

3.1 Particle size characteristics

Samples were taken from the core using 8 cm^3 containers at approximately 10 cm intervals. These samples of approximately 50 g were dried at 60°C and then sieved through a 100 μm mesh. The >100 μm fractions were used for the planktonic foraminiferal species *Globigerinoides ruber* selection (Wang et al., 1999). The <100 μm fractions were then sieved through a 20 μm mesh to separate the < 20 μm fine-size fractions and identify their contents. Only the well-preserved and continuously-deposited sediments were selected and analysed in this study.

Airborne dust is a heterogeneous mixture of particles with variable diameters, varying from 0.1μm to ~1000μm (e.g. Lee et al., 2010). The dust mobilization process shows that only dust particles finer than 20μm can be transported in long-term suspension over a long distance (Pye, 1989; Maher et al., 2010). Palaeo-dust records of global dust sources also show that most of long-travelled dust has similar particle size distribution with size fractions generally less than 20 μm (Arimoto et al., 1997; Reid et al, 2003; Maher et al., 2010). This indicates that the < 20 μm size fraction is a good substitute for long-transported dust fraction (e.g. Sun, 2002). The drilling site of Core G6-4 is ~850 km away from the Australia continent on the down-wind path, indicating dust particles in this core are transported mainly in long-term suspension. Therefore, in order to avoid the effects of particle size on the geochemical contents, only the < 20 μm fraction was used in this study.

3.2 Leaching procedure

Based on age distribution (see Wang et al., 1999), a subset of 25 fine-grained

samples (<20 μm fractions) covering the last 300 ka was selected for Sr-Nd isotope analyses by thermal ionization mass spectrometry (TIMS), and trace elements analysis by ICP-MS.

As Sr is usually significantly enriched in marine carbonates derived from seawater, the Sr isotopic compositions of marine sediments are often seriously affected by seawater Sr and cannot reflect the information of the source materials (Wang et al., 2007). To eliminate this influence, the bulk samples were leached using 1 M HCl as described by Innocent et al. (1999) and Steinmann et al. (2012) for deep-sea sediments. The residues were rinsed three times in distilled water and further treated in sealed teflon beakers with 1 M ammonia to dissolve biogenic opals. After leaching, the remaining sample material (silicate fractions) was dried in an oven at 60°C for three days, and then weighted. The residue weight is considered to represent the weight of the mineral fraction, as any remaining organic matter accounts for less than 1–2%.

3.3 Chemical and isotope analyses

About 50 mg splits of the dried residues were dissolved with a mixture of concentrated HF+HNO_3 solution. After complete digestion and conversion to nitrates, the samples were re-dissolved in 10ml 10% double-distilled HNO_3 to ensure complete homogenization. Then a small precisely weighed aliquot of each sample solution was taken for trace element analysis by inductively coupled plasma mass spectrometry (ICP-MS), and the remaining solution dried down for column chemistry and Sr-Nd

isotope analysis by thermal ionization mass spectrometry (TIMS). The ICP-MS trace element and TIMS Sr and Nd analytical procedures are described in Li et al. (2006) and Uysal et al. (2007).

In the TIMS procedure, Sr and Nd were separated by conventional ion exchange techniques, and then analysed for their isotopic compositions on a VG Sector-54 TIMS. The total procedural blanks in our laboratory were <50 pg for Nd and <100 pg for Sr, which are negligible. $^{87}Sr/^{86}Sr$ ratios were normalized to $^{86}Sr/^{88}Sr = 0.1194$, and $^{143}Nd/^{144}Nd$ ratios were normalized to a $^{146}Nd/^{144}Nd$ ratio of 0.7219. During the course of this study, repeated analysis of the SRM-987 standard (after every 9 samples) gave a mean $^{87}Sr/^{86}Sr$ value of 0.710249±0.000020(2σ) and analysis of the La Jolla Nd standard yielded a mean $^{143}Nd/^{144}Nd$ ratio of 0.511867±0.000012(2σ).

The small aliquot for ICP-MS trace element analysis was dried and re-dissolved in 2% quartz distilled nitric acid which was doped with 6 ppb ^{6}Li-^{84}Sr-^{115}In-^{147}Sm-^{169}Tm-^{187}Re-^{209}Bi-^{235}U internal standards with a typical dilution factor of 1:5000. The dilute solutions were measured on a Thermo X-series quadrupole ICP-MS, also in the Radiogenic Isotope Facility, using a protocol modified after Eggins et al (1997). Instrument response was calibrated against W-2, an international rock standard, and cross-checked with other international rock standards, such as BIR-1 and BHVO-1, which were run as unknowns at the beginning and end of the analytical sequence. External drift correction was performed by repeated analyses (measured after every five samples) of a “drift-monitor" of similar matrix for correction of the instrument’s long-term drift. Our ICP-MS analytical

protocol was based on the use of multiple-element internal standards for mass response corrections, and ^{147}Sm and ^{169}Tm were used as internal standards to ensure high-precision measurements of rare earth element (REE) contents. Appropriate interference corrections are applied to correct for isobaric and molecular species following determination of the oxide and hydroxide production rates from single element interference check solutions. Precisions for most elements in these analyses were routinely better than ±3%, as shown in our repeated measurements of rock standards of different types (see Li et al., 2006).

4. Results

4.1 Trace element geochemistry

As shown in Supplementary Table 1 and Figures 2 and 3, the studied samples exhibit enrichment of light rare earth element (LREE) with La_N/Sm_N (subscript N indicates chondrite normalized value) = 2.5 to 5.1 and slight enrichment of heavy REE (HREE) with Gd_N/Yb_N <1.0 (with the exception of XW72 and XW90, with Gd_N/Yb_N>1.0). As shown in Fig.2, Such a U-type REE pattern is different from those of aeolian dusts and fine-grained sediments from China and Australia with much higher total REE concentration and slight depletion of HREE as evidenced by higher Gd_N/Yb_N ratios (1.4 to 2.1) (Fig.2). Comparing with the Post-Archaeon Australian Shale Composite (PAAS) (Nance and Taylor, 1976; McLennan et al., 1983; Taylor et al., 1986; Pourmand et al., 2012), the studied samples are characterised by low total

REE concentration and strong enrichment of HREE.

All samples display week negative Eu anomalies with Eu anomalies values varying from 0.68−0.85, mostly focusing on 0.70−0.80 (Supplemental Table 1, Fig.2). The prominent feature of these samples is that some samples significantly depleted in Ce (with negative anomalies low to 0.22). Because the net addition of REE to the oceans has no Ce anomaly, these negative Ce anomalies may be attributed to fish teeth and other phases that inherit REE from seawater or presence of Ce-depletion arc volcanic rocks in their provenance (Plank et al and Langmuir, 1998). All samples displayed positive Th, Li, Zr-Hf, and Cu anomalies in the upper continental crust normalized trace element distribution patterns (Fig.3). These samples have lower compatible element concentrations.

4.2 Sr and Nd isotopic compositions

Nd and Sr isotope data are reported in Supplementary Table 2. $^{143}Nd/^{144}Nd$ values in the core vary between 0.512128 and 0.512616, corresponding ε_{Nd} ranging from -0.39 to -9.91. The measured $^{87}Sr/^{86}Sr$ ratios vary from 0.707055 to 0.715033. All the values fall in the range of the India Ocean sediments in general ($^{143}Nd/^{144}Nd$: 0.5119-0.5127, $^{87}Sr/^{86}Sr$: 0.706-0.730) (e.g. Ben Othman et al., 1989; Vroon et al., 1995; Gasparon and Varne, 1998).

Overall, the fine-grained sediments have a narrow range of $^{147}Sm/^{144}Nd$ ratios (0.1051-0.1364), most of which fall in between 0.105 and 0.130. This range is closer to the average value for the continental crust, as reported for ancient clastic sediments

and particulate loads of modern rivers (e.g. Goldstein et al., 1984; Gallet et al., 1996) (Fig.4). The average $^{147}Sm/^{144}Nd$ ratio of the 25 analysed samples is 0.117, indistinguishable from the average value for river and dust particulates in Australia (Fig.4).

Sr-Nd isotopic ratios are nearly constant for samples older than 180 ka, with $^{143}Nd/^{144}Nd$ ratios ranging between 0.512190 and 0.512248 and $^{87}Sr/^{86}Sr$ values ranging between 0.711838 and 0.713273 (Fig.5). In contrast, Sr-Nd isotopic ratios are variable in samples younger than 180 ka (Fig.5). At least 3 elevated $^{143}Nd/^{144}Nd$ values (at 5.4 ka, 22.3 ka, 76.7 ka) can be recognized during the past 180 ka, with radiogenic values peaking at 0.512616, 0.512393, 0.512443, respectively, corresponding to 3 less radiogenic Sr isotopic ratios at 0.707055, 0.709857, 0.708678, respectively. It is worth noting that, excluding these anomalous samples, the post-180 ka section has lower $^{143}Nd/^{144}Nd$ and higher $^{87}Sr/^{86}Sr$ ratios in general than the pre-180 ka section (Fig.5).

The measured Nd and Sr isotopes tightly correlated with Tb/Yb ratios (correlation coefficient, r = 0.85 and 0.76, respectively) and broadly correlated with La/Ni (r = 0.75 and 0.61, respectively) (Fig.6). The samples with high ε_{Nd} and low $^{87}Sr/^{86}Sr$ ratios generally possess high Sm/Nd ratios and contain high dust percentage. For example, the isotopic anomaly of XW-72 at 76.7 ka is correlated with corresponding anomalous values of fine-grained fractions (< 20 μm) (56.54%) and high Sm/Nd ratio (0.23) (Fig.4).

4.3 Sedimentation rate, fine fraction percentage, and Australian dust contribution

The curve of sedimentation rate in Fig.5 was determined from the chronology reported by Wang et al., 1999. Sedimentation rate varies from 1 cm/ka to 9 cm/ka, with an average of 4 cm/ka. Compared with the 180-300 ka section, sedimentation rate becomes more variable in the last 180 ka, especially in the last 100 ka (Fig.5). Fine fraction (<20 μm) percentage shows little significant trend through time, and averaged around 40%. Australian dust contribution in studied samples determined by simple two-component modeling in this study ranges from 8% to 75%, with an average of 60%, and shows a slight increase in the post-180 ka section (Fig.5).

5. Discussion

5.1 Contribution of Australian aeolian dusts

Dust provenance has been constrained most successfully by Nd and Sr isotope systematics (e.g. Dasch, 1969; Nakai et al., 1993; Biscaye et al., 1997; Chen et al., 2007; Hyeong et al., 2011). The Nd isotope compositions of the fine-grained mineral components in Core G6-4 (Fig.4) are slightly more radiogenic than those of Chinese loess sediments (e.g. Gallet et al, 1996; Jahn et al., 2001), which are considered as representative of the average upper crustal values (e.g. Taylor and McLennan, 1985; Rudnick and Gao, 2003). The Sr and Nd isotopes of these samples are also significantly more radiogenic than the Australian Proterozoic Crust (Fig. 4).

As shown in Fig.6, the correlations between Sr-Nd isotopes and trace element

ratios of La/Ni and Tb/Yb suggest two end-member components for the fine-grained samples in Core G6-4. The high-Sr and low-Nd end-member is characterised by ε_{Nd}=-5 to -10, $^{87}Sr/^{86}Sr$=0.710-0.715, La/Ni=0.1-0.6, Tb/Yb≈0.18, which compares well with the composition of aeolian dusts from Australia (Fig.6 and Supplemental Table 3). By contrast, the low-Sr and high-Nd end-member is characterised by ε_{Nd} >0, $^{87}Sr/^{86}Sr$<0.707, La/Ni>0.8, Tb/Yb>0.24, typical characteristics of felsic arc volcanic ashes in Indian Ocean (e.g. Handley et al., 2008). Therefore, the fine-grained samples in Core G6-4 most likely reflect mixing between aeolian dusts from the Australian continent and volcanic ashes from Sunda arcs (Fig.6). This is consistent with the previous investigations of marine sediments in west Sunda arc region (e.g. Gasparon and Varne, 1998; Gingele et al., 2001) and the pollen records of Core G6-4 (e.g. van der Kaars, 1991; Wang et al., 1999).

However, it is difficult to quantify the proportion of each component, mainly because aeolian dusts by the northwest dust path from Australian continent to the Indian Ocean are poorly constrained (e.g. Hesse and McTainsh, 2003; Gingele et al., 2007). Because the different Australian crustal domains have distinctive Sr-Nd isotope compositions (e.g. Grousset et al., 1992; Revel-Rolland et al., 2006; Gingele et al., 2007; Carson, 2013), the estimated average value has not representative meanings. By contrast, average geochemical and Sr-Nd isotopic values of river (e.g. Goldstein et al., 1984; Marx et al., 2005; Gingele et al., 2007; Marx et al., 2009) and aeolian particulates (e.g. Marx et al., 2005) as well as loess (e.g. Gallet et al., 1996; Jahn et al., 2001; Marx et al., 2005; Marx et al., 2009) generally used to as an index of average

upper continental crust. However, it is unclear that such values are also representative of mean aeolian dusts derived from the Australia continent since the Australian continent may be significantly different from the rest of the world (Drummond and Collins, 1986).

Due to the prevailing easterly winds in the region, we assume that arid areas in western, northern, and central Australian regions could be potential dust sources for the eastern Indian Ocean (McTainsh, 1989; Hesse and McTainsh, 2003; Stuut et al., 2014). It has been documented that the Lake Eyre Basin of central Australia is the most active dust storm region on the continent, being capable of affecting most of Indonesia many thousands of kilometers from the Australian continent in a relatively short period of time (McGowan and Clark, 2008). Although Western Australian deserts consist mainly of sand dunes, they are not considered to be a major atmospheric source for long-transport dusts (Prospero et al., 2002) since they are characterised by low dust entrainment, great geomorphic stability and enrichment in larger particles with very high settling velocity (Prospero et al., 2002; Hesse and McTainsh, 2003). Thus, considering that Northern Territory is close to the core location, it appears that the major sources of dusts transported to Core G6-4 are from Northern Territory in northern Australia and Lake Eyre Basin in central Australia.

Overall, Sr-Nd isotopic data on potential dust sources in Australia are scant and patchy. Previous studies reveal a wide range of Sr and Nd isotopic ratios in these regions: Northern Territory in northern Australia with $^{87}Sr/^{88}Sr$ = 0.715-0.732 and $^{143}Nd/^{144}Nd$ = 0.5118-0.5120 (Revel-Rolland et al., 2006); Great Sandy Desert in

Western and Central Australia with $^{87}Sr/^{86}Sr$ =0.732-0.763 and $^{143}Nd/^{144}Nd$ =0.512(Grousset et al., 1992); Lake Eyre Basin in central Australia with $^{87}Sr/^{88}Sr$ = 0.7090-0.7098 and $^{143}Nd/^{144}Nd$ = 0.5124-0.5125 (Revel-Rolland et al., 2006). Considering the contributions of dust sources, the first 8 samples in Supplementary Table 3 (4 from Lake Eyre Basin, 3 from Northern Territory and 1 from Great Sandy Desert) were used to identify average isotopic ratios, which are around 0.720 for $^{87}Sr/^{86}Sr$ and 0.5120 for $^{143}Nd/^{144}Nd$. We use these data to define the Sr-Nd isotopic field for Australia dusts with $^{87}Sr/^{86}Sr$ ratios ranging from 0.709 to 0.731 and ε_{Nd} = -3 to -15 (Revel-Rolland et al., 2006).

In contrast, the isotopic ratios for volcanic rocks from the Sumatra-Salak-Java-Bali-Sumbawa Islands adjacent to Core G6-4 are less variable, overlapping with the field of the Indian Ocean sediments as well (e.g. Ben Othman et al., 1989; Gasparon and Varne, 1998) (Fig.4). The published isotopic data are summarized in Figure 4 and compiled in Supplementary Table 3, which shows $^{87}Sr/^{86}Sr$ ratios ranging from 0.704 to 0.708 with an average around 0.7065, and $^{143}Nd/^{144}Nd$ ratios varying between 0.5125 and 0.5129, with an average around 0.5127 (Whitford et al., 1981; Ben Othman et al., 1989; Gasparon and Varne, 1998; Turner and Foden, 2001; Gertisser and Keller, 2003; Handley et al., 2008; Sendjaja et al., 2009).

The isotopic data of volcanic ashes from the region closest to Core G6-4 were reported by Ben Othman et al. (1989). These authors analysed a volcanic sample from a water depth of 3230m, south of the Sumba Island (9º28´S, 122º21´E) and obtained a

value of 0.705853 for $^{87}Sr/^{86}Sr$ ratio and 0.512734 for $^{143}Nd/^{144}Nd$. Therefore, the isotopic values of 0.706-0.707 for Sr and 0.5127 for Nd are considered to be most representative of volcanic ashes mixing into the core sediments.

The published Sr and Nd isotope data of volcanic ashes from nearby volcanoes, aeolian dusts from Australian continent are used to identify the potential-sediment source of the samples from Core G6-4 (Fig.4). Volcanic ashes are characterised by radiogenic Nd isotope composition ($^{143}Nd/^{144}Nd>0.5125$ in most samples) and less radiogenic Sr isotope composition ($^{87}Sr/^{86}Sr<0.707$ in most samples). Aeolian dusts from Australian continent have homogeneous, less radiogenic Nd and highly radiogenic Sr isotope compositions (with $^{143}Nd/^{144}Nd<0.5125$ and $^{87}Sr/^{86}Sr>0.708$). As shown in Fig.4, the Sr-Nd isotope compositions of fine-grained samples from Core G6-4 plotted along the mixing trend between arc volcanic ashes and aeolian dusts from Australian continent. This suggests a mixed provenance for these sediments.

A two-end-member mixing model was performed according to the follow end-parameters (see details in Supplementary Table 4). Volcanic ashes have Nd = 10 ppm, Sr = 500 ppm (Lytwyn et al., 2001; Turner and Foden., 2001), $^{143}Nd/^{144}Nd$ = 0.5127 and $^{87}Sr/^{86}Sr$ = 0.706-0.707, and continental dusts have Nd = 32 ppm Sr = 100 ppm (Kamber et al., 2005; Marx et al., 2005;Gingele et al., 2007), $^{143}Nd/^{144}Nd$ = 0.5120-0.5121 and $^{87}Sr/^{86}Sr$ = 0.720.

As shown in Fig.4, the mixing lines bracket the data array very nicely. The two-end member mixing modeling suggests that the majority of the core samples contain 50-80% aeolian dusts derived from the Australian continent. This is consistent

with pollen records from this core (Wang et al., 1999) and the Argo Abyssal Plain core G6-2 (van der Kaars, 1991) that show that Australian dust contributes the most significant part to the sediments in Eastern Indian Ocean, as evidenced by the occurrence of large amounts of chenopod pollen grains in the core sediments (van der Kaars, 1991).

5.2 Australian continental aridity: a possible contributor to the secular isotopic change

After eliminating the anomalous values that may be indicative of ash inputs from short episodes of local volcanic eruptions as will be discussed in Section 5.3, the mean $^{143}Nd/^{144}Nd$ prior to 180 ka is 0.51225, increasing to 0.51215 in the post-180 ka section, whilst $^{87}Sr/^{86}Sr$ shows an opposite trend from 0.7125 to 0.7140. Overall, sediments older than ~180 ka are more radiogenic in their Nd isotope compositions and less radiogenic in their Sr isotope compositions than afterwards (Fig.5).

In general, there are three possible interpretations for the secular change of $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios at ~180 ka: provenance change, chemical weathering change (i.e. aridity, stronger winds), or grain-size distribution change (Hesse, 1994; Wang et al., 2007; Meyer et al., 2011). Because we did not observed the major change in grain size in Core G6-4 (Fig.5 and Supplementary Table 2), the possibility of major grain-size distribution change can be ruled out. Furthermore, as shown in Fig. 5, the proportion of fine fraction in studied samples displays no meaningful correlation with the total REE, $^{87}Sr/^{86}Sr$ ratio and ε_{Nd} (Fig.7), suggesting that grain-size distribution is

not likely to be an important controlling factor for the Sr-Nd isotopic change. The studied samples display a narrow range of the abundance of fine fraction (from 25% to 55%), which should be related to the strong heterogeneity of both aeolian dust and volcanic ashes.

Sedimentation rate is another possible factor that influences the geochemical and isotopic changes (e.g. Wang, 1999). However, sedimentation rate in the studied samples show little correlation with the total REE, $^{87}Sr/^{86}Sr$ ratio and ε_{Nd} (Fig.7), indicating that sedimentation rate is not likely to be a controlling factor for the secular Sr-Nd isotopic change. The extremely low sediment rate during marine isotope stage (MIS) 3 is related to the poor representation of MIS 3 in the chronology (Wang et al., 1999). Although the reason for the missing of MIS 3 is not clear, such hiatus in the record could not influence the secular geochemistry isotopic pattern as there is no geochemical and isotope dates located in this stage. The variability in sedimentation rate in the last 100 ka can be partly attributed to large sampling intervals for oxygen analysis (more than 10 cm in this study), possible biological mixing events and disturbances by volcanic eruptions (Hess, 1994).

In addition, it is unlikely that this secular Sr-Nd change was caused by stronger wind strength because, compared with MIS 6 (195-128 ka), the glacial MIS 8 (301-244 ka) with similar strong winds did not see corresponding Sr-Nd isotopic change. However, it is worth noting that some other factors, such as small changes in provenance, cannot be entirely ruled out based on current available data.

A marked increase in dust supply usually occurs in response to a general increase in aridity (e.g. Hess, 1994; Hess and McTainsh, 2003; Marx et al., 2009). The dust contribution shows a slight increase in the post-180 ka section, matching with the secular Sr-Nd isotopic change (Fig.5). Thus, we consider that the secular Sr-Nd isotopic change may be attributed to Australian continental aridity.

Drier periods, especially the periods with increased aridity, are indicated by low Eucalyptus, Woodland group and high Gramineae and Grassland Group. In the pollen records of Core G6-4, there are sharp declines in Eucalyptus and Woodland group percentage at approximately 460 cm core depth with an interpolated age of ~180 ka (Fig.8). Conversely, the strong increases in the Grassland group and Gramineae at ~180 ka can also been observed (Fig.8). Charcoal record may also complement pollen records in indicating the degree of aridity (e.g. Wang et al., 1999). Although there is a great deal of variation in charcoal record through the record, an overall increased pattern is clearly discernible (Fig.8). After ~180 ka, open grasslands were the dominant vegetation, and only during the few wetter intervals shrubland and open woodlands with eucalypts were present, apparently showing that a major vegetation change took place at ~180 ka (Fig.8). The development of a much more open vegetation type is suggestive of a drier climate, indicating that the climate might become much drier in Australia since ~180 ka.

High aeolian flux of dust in two cores (C1/86 6GC3 and E39.75) from Tasman Sea sediments began early in MIS 6 (～185 ka) (Hesse, 1994) (Fig.8), which is coincident with the timing of the increase of Australian dust input in this study. In

addition, increased aridity since 180 ka also appears to be found in palynological records from northern Australia (Kawamura et al., 2006) and the well-dated fauna records from Mt Etna, central Queensland as described above(Hocknull et al., 2007). All these studies confirm that the transition to true aridity in Australia could start from MIS 6 which was further intensified in the subsequent glacial periods.

Therefore, the good match between the secular Sr-Nd isotopic change and the timing of increased Australian continental aridity suggests that Australian continental aridity is a possible contributor to the secular Sr-Nd isotopic change. Yet, it cannot be determined whether Australian continental aridity is the only essential factor because even small changes in dust provenance or the combination of the other factors mentioned above can also cause such a degree change in Sr-Nd isotopic compositions.

5.3 Correlation with Sunda arc volcanicity

The Sunda-Banda arc system is well known for its spectacular volcanic activity, including the 1883 eruption of Krakatau, Java, the 1815 eruption of Tambora, Flores. Intriguingly, there are few long-term records of earlier, larger Quaternary eruptions from Southeast Asia. What is more, volcanic eruptions, especially large events, are an important natural cause of climate change as they inject huge volumes of ash and gases into the atmosphere (Robock, 2000), indicating the identification of pre-historic of volcanic eruptions can provide the underpinning data for a better understanding of important radiative and dynamical processes that respond in the climate system to both natural and anthropogenic forcings.

In this study, three anomalies (at 5.4, 22.3 and 76.7 ka) characterised by elevated $^{143}Nd/^{144}Nd$, decreased $^{86}Sr/^{87}Sr$ and extremely low Australian dust contribution occur in the last 180 ka (Fig.5). They are obvious evidence for volcanic eruptions in the nearby volcanoes, since no continental input can generate such a degree of anomaly (note that the three data points plot entirely within the Banda and Sunda arc volcanic field in Fig. 4). The other three samples XW41, XW51, XW62 (at 119,148 and 192 ka) also show some degree of abnormal, however, their origin is unclear. They may be related to either volcanic eruptions or heterogeneity in the dusts.

The third isotopic anomaly (76.7 ka) matches well within its age uncertainty with the Late Pleistocence ~75 ka eruption of the Younger Toba Tuff (YTT) that is the largest known Quaternary eruption, indicating that isotopic anomalies in deep sea sediment cores are good indicators of past volcanic eruption events. The ~75 ka eruption of Toba is thought to have expelled 2800 km^3 of material, 800 km^3 of which was deposited as ash fall (Rose and Chesner, 1990; Oppenheimer, 2002; Lane et al., 2013), triggering a long-term climatic cooling of one degree and even a population bottleneck in the fauna records indicative of mass extinction (Ambrose et al., 2003; Gathorne-Hardy and Harcourt-Smith, 2003). In our record, we identify another two large volcanic eruptions at around 5 ka and 25 ka, although their potential impact on late Quaternary climate is unclear. Our record also implies the likelihood that volcanism along the Sunda-Banda Arc system may become more active in the last 80 ka.

6. Conclusions

Geochemical and Sr-Nd isotopic compositions indicate that fine-grained detrital sediments in Core 6-4 were derived by mixing between aeolian dusts from the Australian continent and volcanic ashes from Sunda arcs. Based on the isotopic data ($^{87}Sr/^{86}Sr$ ratios ranging from 0.707 to 0.715 and $^{143}Nd/^{144}Nd$ between 0.5121 and 0.5126), it is estimated that Australian continental dust input varies from 50 to 80% for the majority of the samples, consistent with pollen records. There is an overall secular change in Sr isotopic ratios from 0.7125 to 0.7140 and Nd isotopic ratios from 0.51225 to 0.51215 post-80 ka, which matches well with the timing of increased Australian continental aridity identified by many other palaeo-environmental records. Although some other factors (e.g. small changes in dust provenance) cannot be entirely ruled out, this good match indicates that Australian continental aridity is a possible contributor to the secular isotopic change. Three isotopic anomalies (5.4, 22.3 and 76.7 ka), characterised by elevated $^{143}Nd/^{144}Nd$ and reduced $^{87}Sr/^{86}Sr$ ratios, occurred in the post-180 ka core section. The third anomalies matches well within its age uncertainty with ~75 ka Toba eruption, and the other two anomalies (at around 5 ka and 25 ka) could be related to another two large volcanic eruptions.

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Figure captions

Figure 1 Map of the Australian/Southeast Asian region showing the location of marine core G6-4 , the distribution of active volcanoes (after Gertisser and Keller, 2003 and Poorter et al., 1991), the major oceanic current patterns (from http://www.indiana.edu/~geol105/1425chap4.htm), the paths of volcanic ashes (see data source in Supplementary Table 2) and the NW aeolian dust path in related to Eastern Indian Ocean (after Bowler, 1976 and Pack et al., 2003).

Figure 2 Chondrite-normalized and Post-Archaean Australian Shale (PAAS) normalized rare earth element patterns for fine-grained samples from Core G6-4. Chondtite values and PAAS values from Sun and McDonough, 1989 and Pourmand et al., 2012 respectively. REE results of aeolian dusts in China are from Gallet et al., 1996, Honda et al., 2004Yang et al., 2007, and REE results of aeolian dusts in Australia are from Petherick et al., 2009 and Marx et al., 2005.

Figure 3 Upper continental crust normalized trace element distribution patterns for fine-grained samples from Core G6-4.

Figure 4 Variation of $^{143}Nd/^{144}Nd$ and $^{86}Sr/^{87}Sr$ for mineral components in the Core G6-4. Date sources: MORB, Indian Ocean: Ito et al., 1987; Rehka¨mper and Hofmann, 1997, OIB, Indian Ocean: Hamelin et al., 1986; Barling and Goldstein, 1990, Banda & Sunda Arc Volcanics: Ben Othman et al., 1989; Della-Pasqua et al., 1995; Vroon et al., 1995; Gasparon and Varne, 1998; Handley et al., 2008; Indian Ocean Sediment: Ben Othman et al., 1989; Gasparon and Varne, 1998; Australian Continental Crust: Grousset et al., 1992; Revel-Rolland et al., 2006; Gingele et al., 2007; Carson, 2013;

Chinese Loess: Gallet et al., 1996; Honda et al., 2004 and Yang et al., 2007; Australian aeolian dusts in the upper panel: Petherick et al, 2009 and Marx et al., 2005; Australian aeolian dusts in the lower panel: Kamber et al., 2005; River & Aeolian Particulates: Goldstein et al., 1984; Australian Early and Middle Proterozoic Crust: Zhao et al., 1992. CHUR: Chondritic Uniform Reservoir; CC: Continental Crust.

The two end-member mixing model is also shown here. Both the red line and the purple line in Fig. 3a are the mixing curves of volcanic ashes the region closest to Core G6-4 and aeolian dusts from Australian continent. The values used in the red line are $^{143}Nd/^{144}Nd=0.5127$, $^{87}Sr/^{86}Sr=0.707$ for volcanic ashes and $^{143}Nd/^{144}Nd=0.5121$, $^{87}Sr/^{86}Sr=0.720$ for aeolian dusts. The values used in the purple line are $^{143}Nd/^{144}Nd=0.5127$, $^{87}Sr/^{86}Sr=0.706$ for volcanic ashes and $^{143}Nd/^{144}Nd=0.5120$, $^{87}Sr/^{86}Sr=0.720$ for aeolian dusts. The concentrations of Sr and Nd isotopic compositions used here are Nd=10ppm, Sr=500ppm for volcanic ashes (Lytwyn et al., 2001; Turner and Foden., 2001); Nd=32ppm, Sr=100ppm for continental crust (Kamber et al., 2005; Marx et al., 2005;Gingele et al., 2007). The data plotted in the figure are also reported in Supplemental Table 3.

Figure 5 Map showing variation of $^{143}Nd/^{144}Nd$, $^{147}Sm/^{144}Nd$, and $^{86}Sr/^{87}Sr$ values as well as sedimentation rate, fine-grained size distribution, Australian dust contribution through time in Core G6-4. The refinement of the oxygen isotope record providing a firmer chronology is from Wang et al., 1999. The average lines (in blue) are also shown in these plots after excluding the three anomalous values. The Australian dust contribution was determined by the two end-member mixing model (see discussion in

Section 5.1), and sedimentation rate was determined using the age model described in Wang et al., 1999.

Figure 6 Plot of Tb/Yb and La/Ni against (a) and (b) ε_{Nd} and (c) and (d) $^{87}Sr/^{86}Sr$ ratio indicate that the fine-grained sediments in Core G6-4 are consistent with a two-component mixing provenance. La/Ni, Tb/Yb, ε_{Nd} and $^{87}Sr/^{86}Sr$ data for volcanic ashes are from Turner and Foden (2001), Handley et al (2008), Sendjaja et al (2009) and Gasparon and Varne (1998), and ε_{Nd} and $^{87}Sr/^{86}Sr$ data are also reported in Supplemental Table 3. ε_{Nd} and $^{87}Sr/^{86}Sr$ data for Australian aeolian dusts are from Grousset et al (1992), Kamber et al., 2005 and Revel-Rolland et al (2006), while La/Ni and Tb/Yb data for Australian aeolian dusts are from Kamber et al., 2005, Max et al., 2005 and Petherick et al., 2009.

Figure 7 Plot of the total REE, $^{87}Sr/^{86}Sr$ ratio and ε_{Nd} against (a), (b) and (c) proportion of fine faction and (d), (e) and (f) sedimentation rate indicate that grain size distribution and sedimentation rate are unlikely to be the main factors for major geochemical and isotopic changes.

Figure 8 Representative palaeo-environmental records in Core G6-4 and some evidences collected from the references indicate that the climate might become much drier in Australia since 180 ka: (a) Eucalyptus record in Core G6-4 (from Wang et al., 1999); (b) Gramineae record in Core G6-4 (from Wang et al., 1999); (c) the percentages of Woodland group in Core G6-4 (from van der Kaars, 1991); (d) the percentages of Grassland group in Core G6-4 (from van der Kaars, 1991); (e) Charcoal concentration values in Core G6-4 (from Wang et al., 1999); (f) $\delta^{18}O$

Globigerinoides ruber values in Core G6-4 (from Wang et al., 1999); (g) aeolian mass accumulation rate in Core E39.75 (from Hess, 1994), blue line is trigger core record, black line is piston core record; (h) aeolian mass accumulation rate in Core C1/86 6GC3(from Hess, 1994).

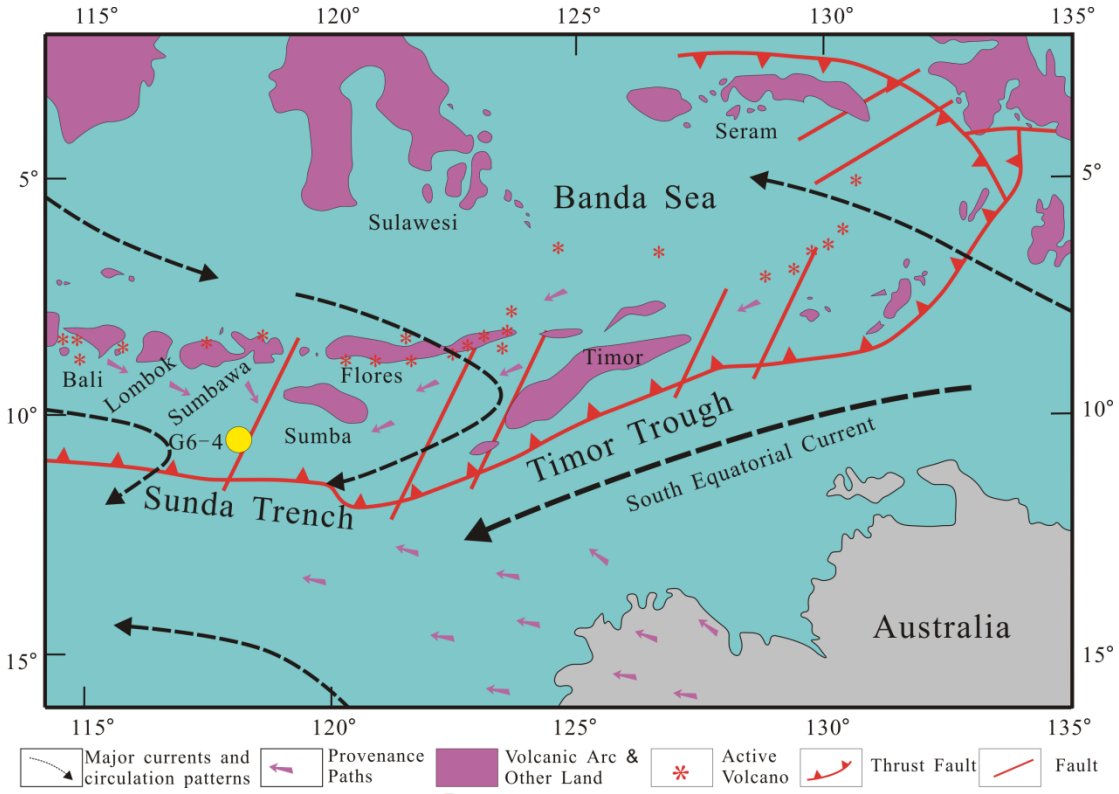

Figure 1

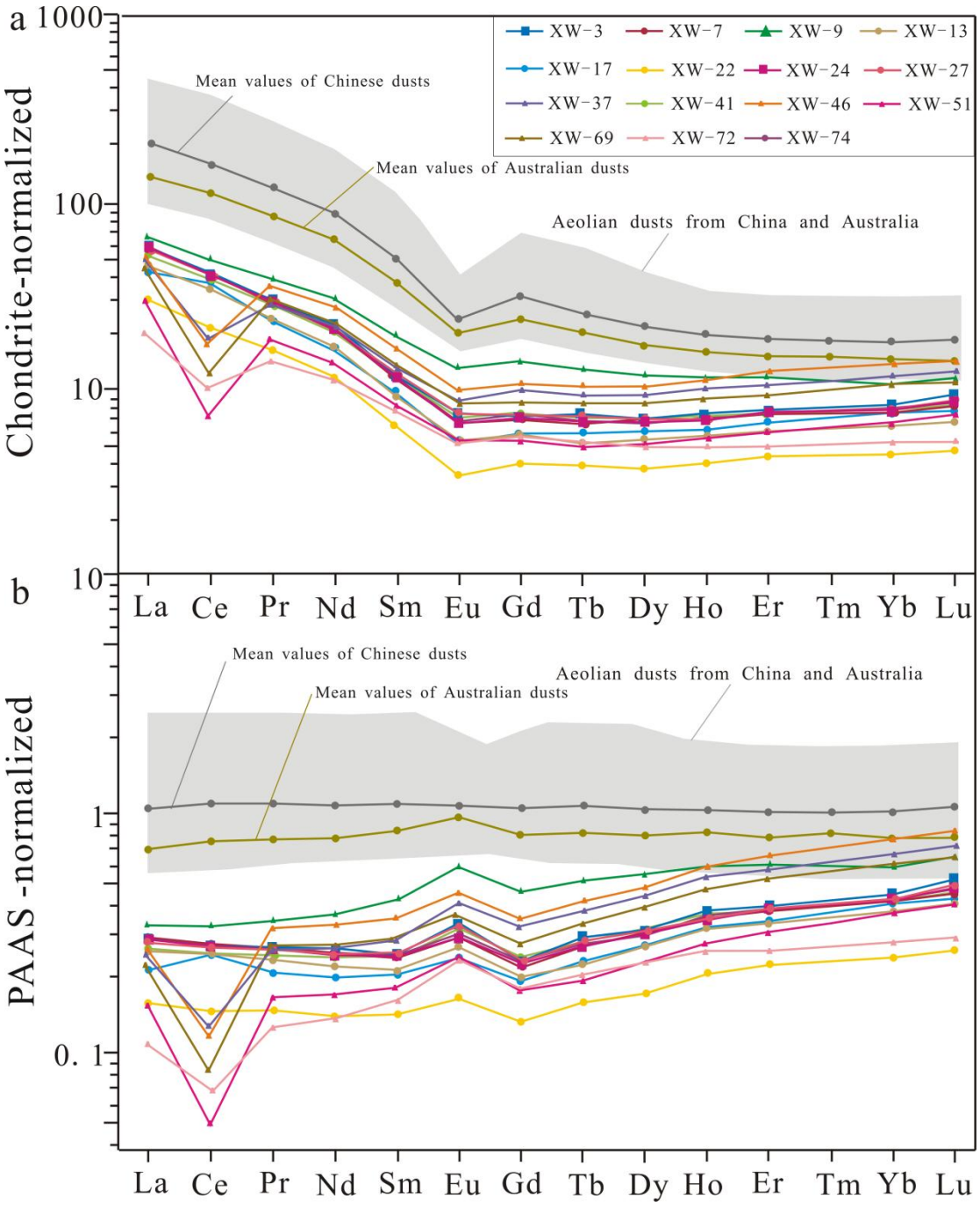

Figure 2

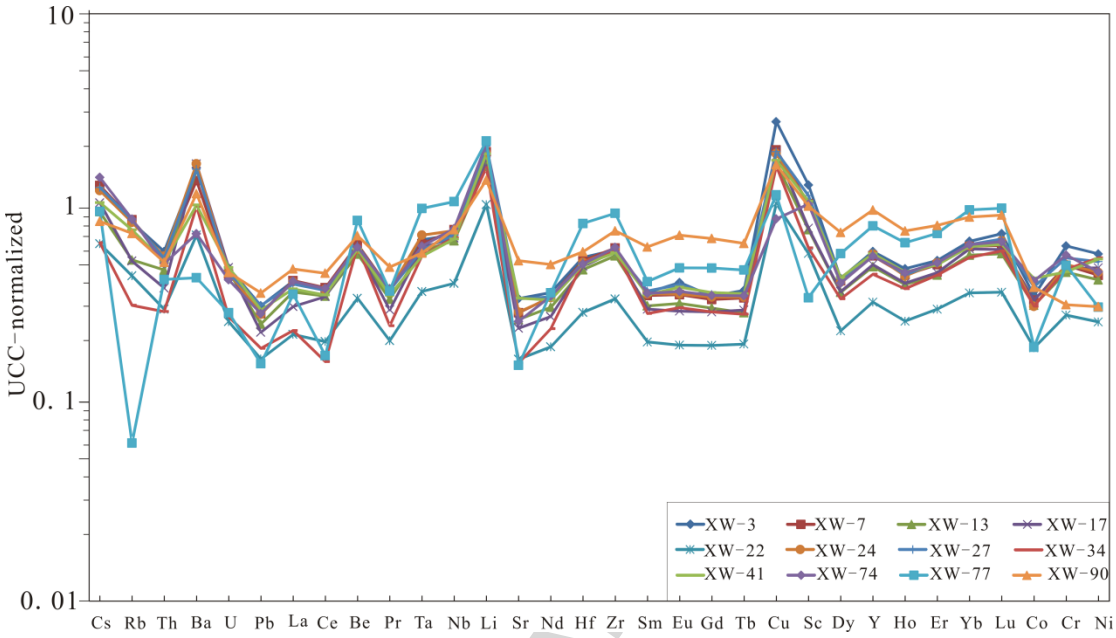

Figure 3

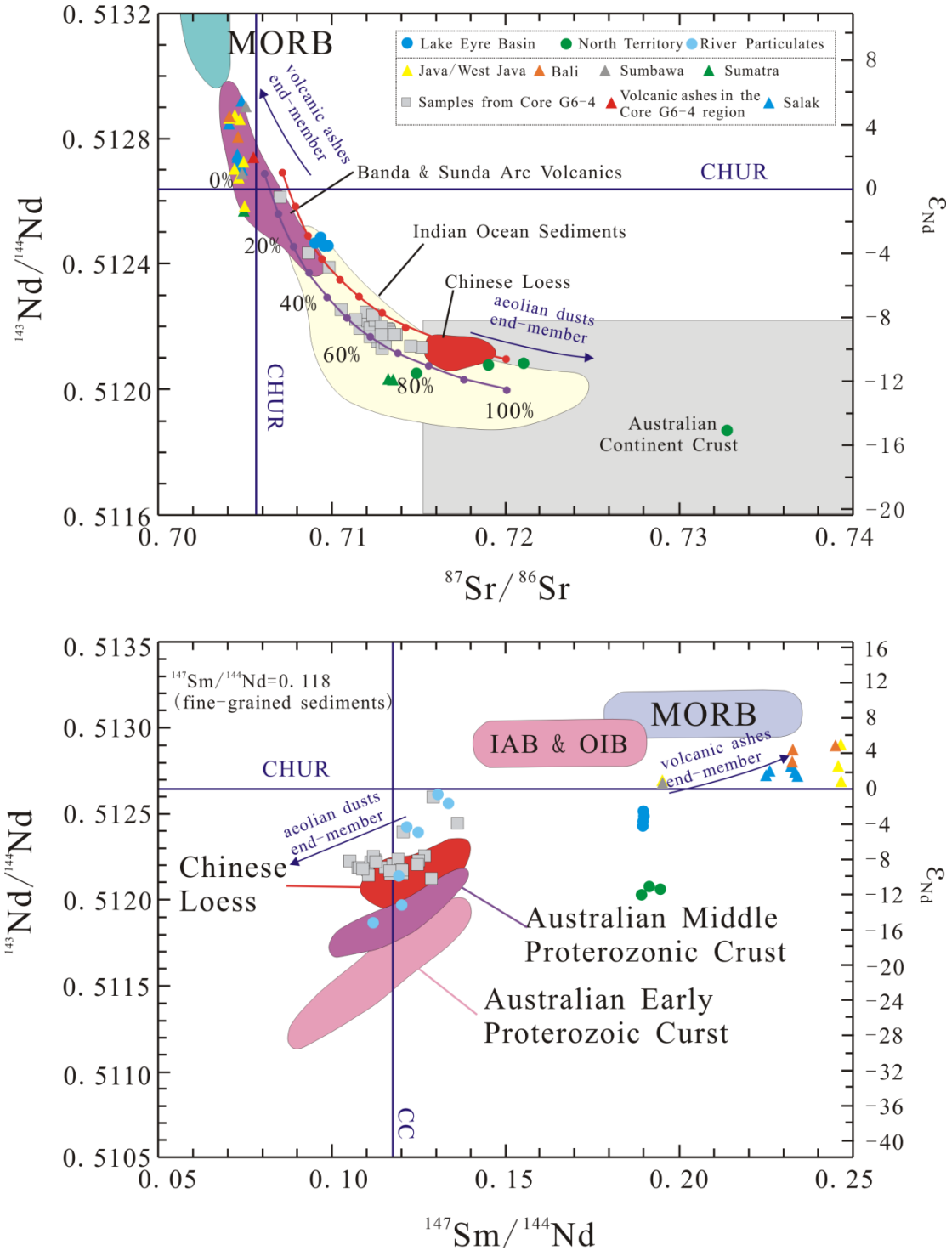

Figure 4

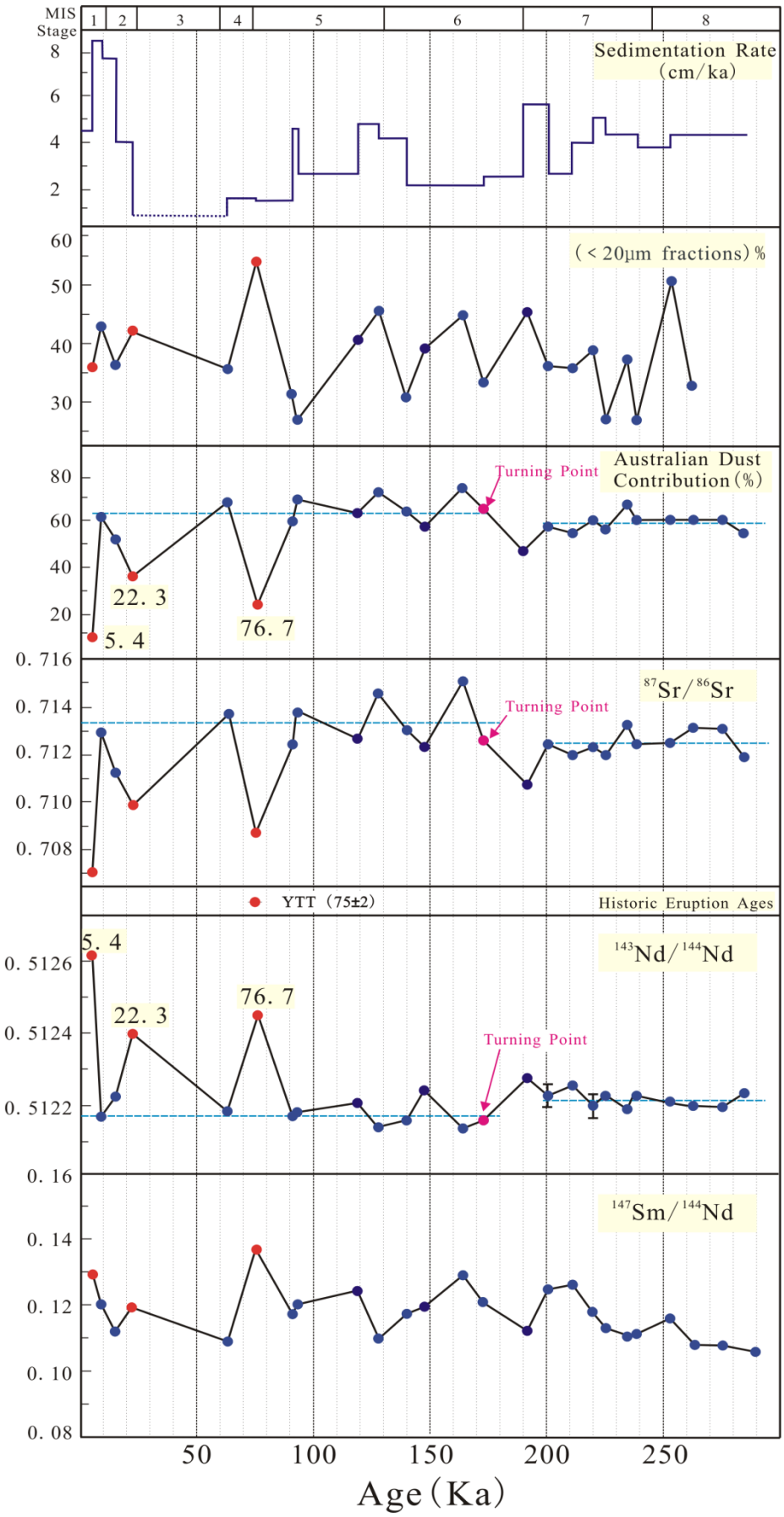

Figure 5

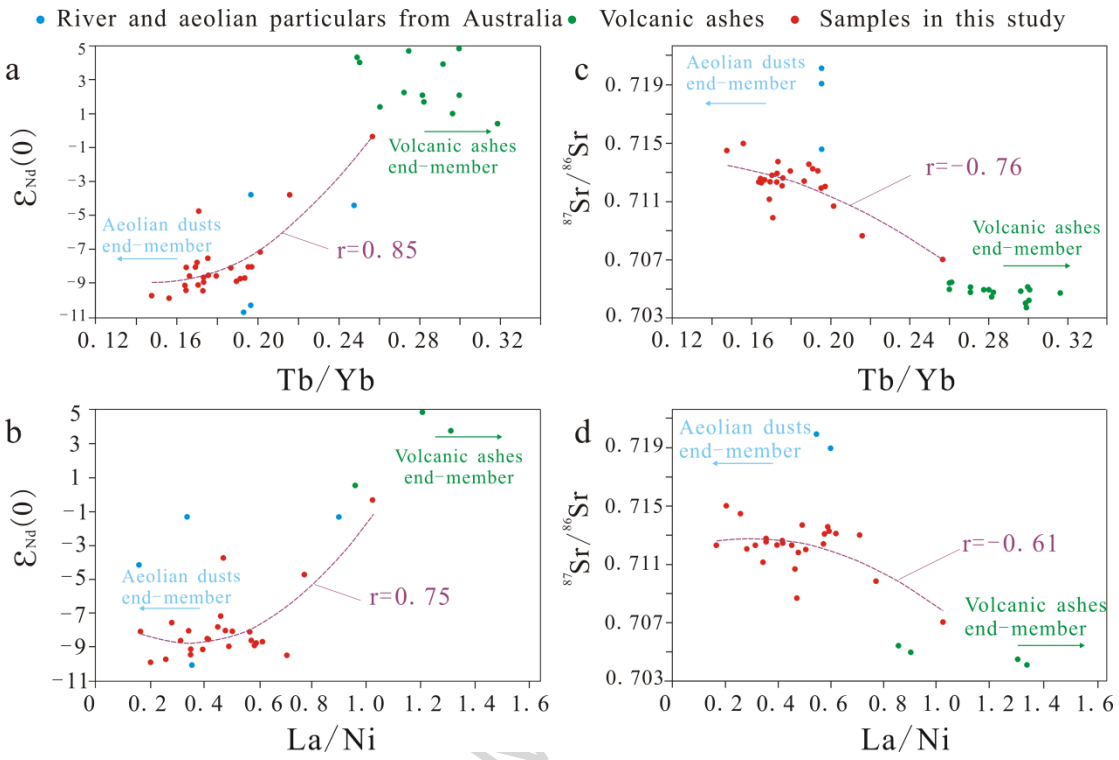

Figure 6

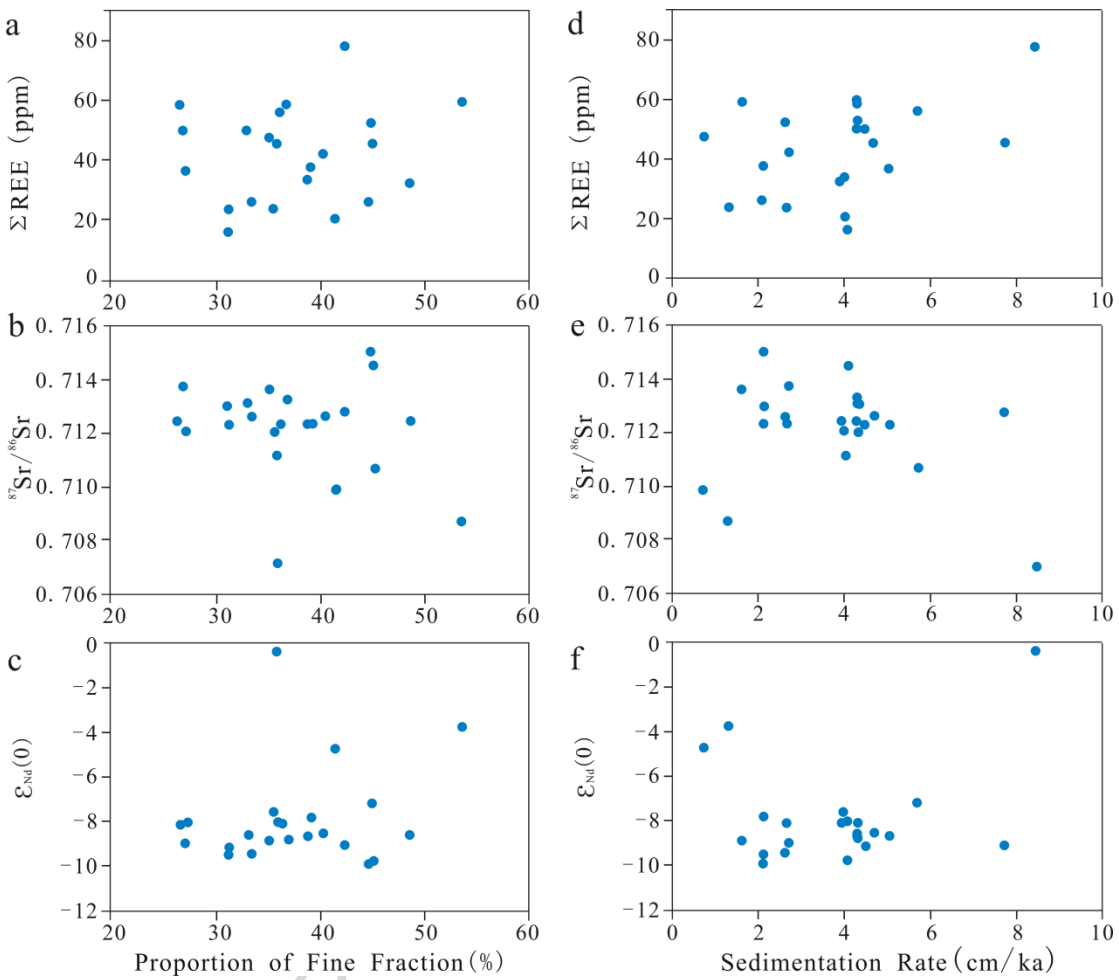

Figure 7

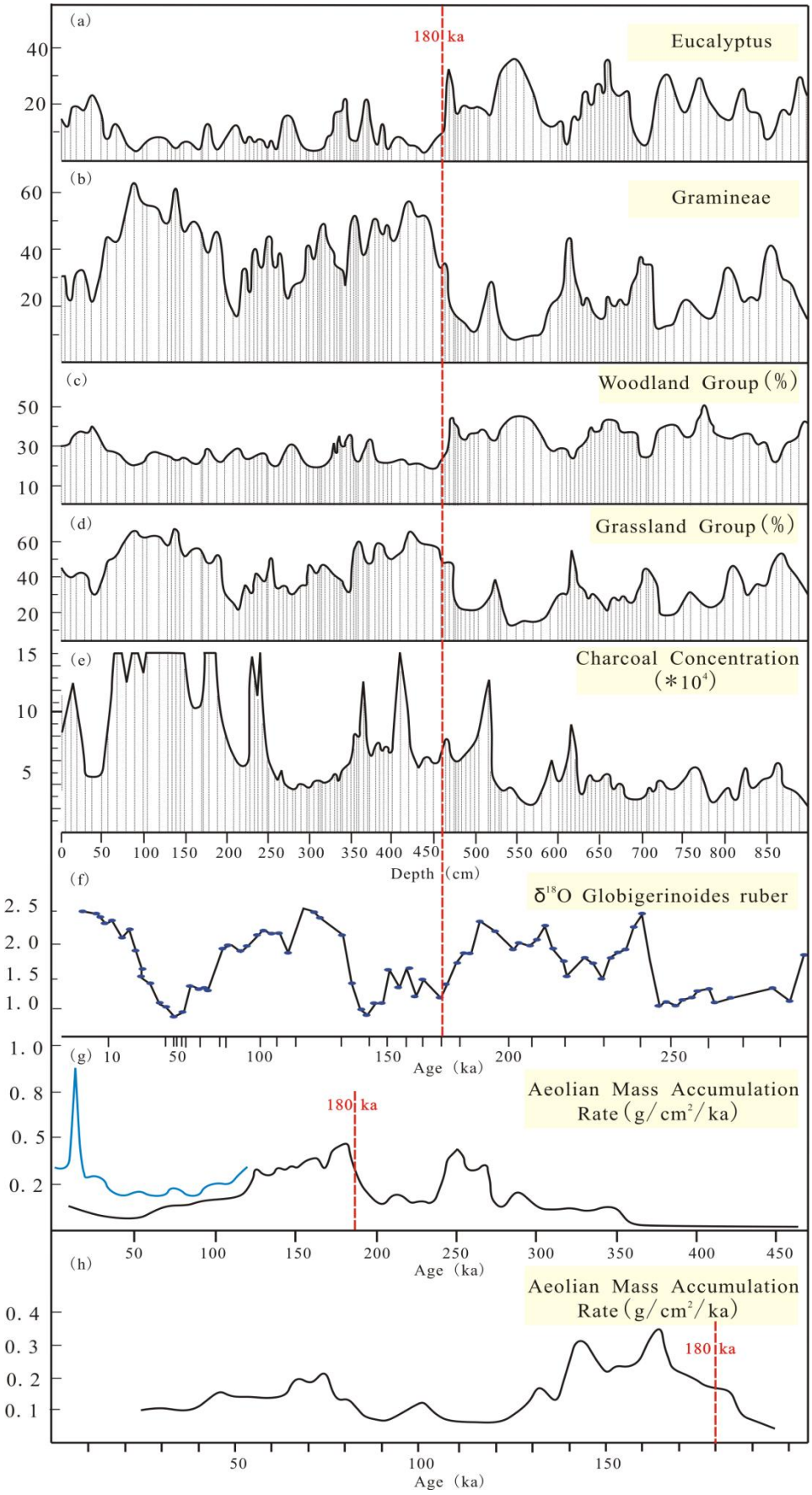

Figure 8

Highlights

- Australian continental dust input varies from 50 to 80% over the last 300 ka
- Australian continental aridity is a possible contributor to the isotopic change
- Three isotopic anomalies (5.4, 22, 77 ka) are related to nearby volcanic eruptions